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Electric Field Induced Transformations and Dielectric Properties in Non-Tilted Phases of a Bent-Core Smectic Liquid Crystal

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We report the structural and relaxation investigations of an achiral bent-core compound in $SmAP_A$ phase. The studied material undergoes transition from one biaxial state to the other via a uniaxial state under electric field. These transformations are explained as successive transition from antiferroelectric to ferroelectric states, where, the intermediate uniaxial state has the molecular secondary directors in the neighbouring smectic layers perpendicular to each other. We also found that near $SmAP_R$ to $SmAP_A$ phase transition the second harmonic electro-optic response, induced polarization, and the dielectric behaviour hint softening behaviour in the material.

Keywords Antiferroelectric order; bent-core molecules; dielectric relaxation; electro-optic effects; polarization

1. Introduction

In 1992, Brad *et al.* [1] suggested a model of ferroelectricity for orthogonal smectic phases. The first experimental evidence of ferroelectricity in non-chiral bent-core materials was reported in 1996 by Niori *et al.* [2]. The orthogonal smectic phases of bent-core materials consist of $SmAP_A$ [3–6] and $SmAP_R$ phases [7]. $SmAP_A$ is a phase in which the polar-directors in the neighbouring layers are arranged anti-parallel and ferroelectricity was induced by the application of electric field. The induction of ferroelectricity in these phases could be after a threshold field or a continuous transition [8]. $SmAP_R$ is a phase wherein inlayer polarization directions

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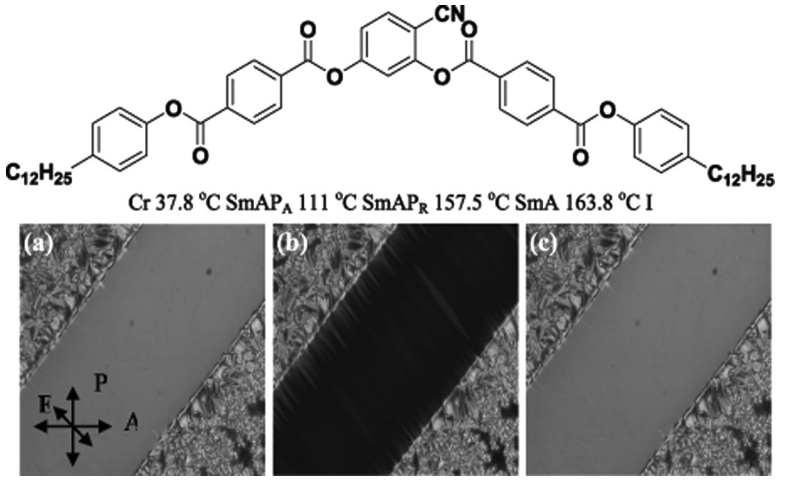

Figure 1. Molecular structure, phase sequence of CK64 and textural transformations in a 9 μm homeotropic cell at 95 °C under **E**, 110 Hz. (a) $\mathbf{E} = 0.8\,\mathrm{V}/\mu\mathrm{m}$, (b) $\mathbf{E} = 1.6\,\mathrm{V}/\mu\mathrm{m}$ and (c) $\mathbf{E} = 2\,\mathrm{V}/\mu\mathrm{m}$.

are degenerated and randomly ordered in the absence of electric field. Recently Shimbo *et al.* have demonstrated a LCD mode out of $SmAP_R$ phase [9].

Phase transitions from SmA to $SmAP_A$ and $SmAP_R$ to $SmAP_A$ (on cooling) have been seen in a number of liquid crystalline materials and their electro-optical, X-ray, dielectric properties have been reported. During paraelectric to biaxial smectic phase transition critical freezing of molecular rotation, a second order phase transition, strong softening of the polar mode in the SmA phase and highly intensive dielectric mode in $SmAP_A$ phase are observed in material with weak antiferroelectric interaction system [10]. In this paper we present results on electric field induced structural transformations in a soft/weak antiferroelectric, polar, biaxial system. Induced polarisation, 2nd harmonic electro-clinic response and dielectric properties of this material hint a softening during transition from $SmAP_R$ to $SmAP_A$ phase.

The studied molecular system CK64 is a first material of its kind which is an example for a continuous growth of polar order of unique phase sequence ($SmAP_A$-$SmAP_R$-SmA) [11]. The molecular formula and phase sequence are given in Figure 1.

2. Experiment

Planar cells (of ITO glass substrates for electro-optic studies and brass plates for dielectric studies) were prepared by coating polymer alignment layer RN1175 (Nissan Chemicals, Japan). Homeotropic cells used in electro-optics were prepared by coating AL60702 (JSR Korea) polymer alignment layer. The electrodes on homeotropic cells were arranged to apply in-plane electric field by etching ITO electrodes on the bottom substrate and the distance between the electrodes was 180 μm. The dielectric response was measured for planar cells of 10 μm thickness using broadband high resolution Alpha-A frequency analyzer (Novocontrol GmbH, Germany).

3. Results and Discussion

3.1. *Textural Transformations*

$SmAP_A$ phase under crossed polarisers shows schleiren texture of $s = 1, 1/2$ declinations.

When an in-plane electric field of 100 Hz up to 0.5 V/μm is applied to a homeotropic cell the schleiren texture transforms to a uniform texture and saturates with biaxiality $\delta n = 0.016$ (Fig. 1b). When the strength of the field increased to 1.6 V/μm, the texture changes to completely dark uniaxial texture ($\delta n = 0$) (Fig. 1c). Further slight increase of voltage brings back the uniaxial texture to a biaxial texture again ($\delta n = 0.016$). The threshold voltages required to achieve these textural transformations is a function of temperature and frequency (Fig. 2 inset).

The transition from a birefringent state to a uniaxial state in $SmAP_A$ is further seen in the 2nd harmonic of electro-clinic response of a homeotropic cell, which is measured using a lock-in amplifier (Stanford Research Systems SR830). A measurement of the response at fundamental frequency gives very low signal while the second harmonic gives a large signal. The voltage dependence of 2nd harmonic response of transmittance for different temperatures in $SmAP_R$ and $SmAP_A$ phases is shown in Figure 2.

The physical phenomena governing this textural transformation could be explained as follows: In $SmAP_A$ phase the minor directors in the same layer are parallel to each other but they are anti-parallel between neighbouring layers. As we have seen above, the response of this structure to the electric field is no longer Langevin but shows three optically distinguishable states. For lower electric field this antiferroelectric arrangement is slightly distorted and for higher electric field it completely transforms into ferroelectric structure. For the intermediate field strength it goes

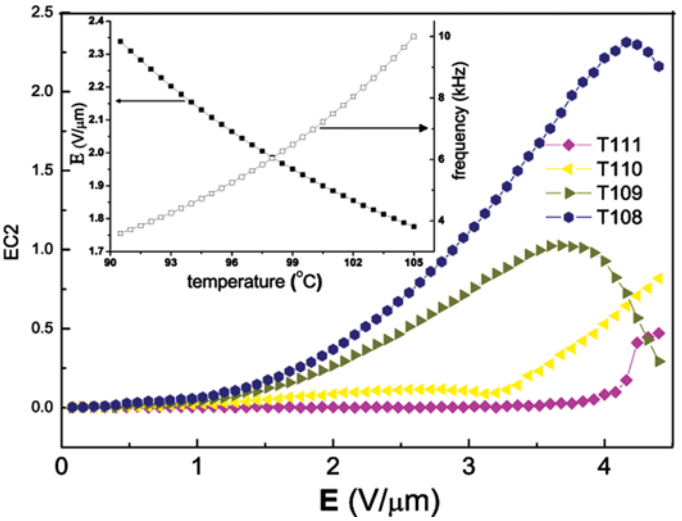

Figure 2. 2nd harmonic electro-optic response with electric field in a 9 μm homeotropic cell at 110 Hz. Inset: Temperature dependence of voltage (of frequency 110 Hz) and temperature dependence of frequency (of $\mathbf{E} = 2$ V/μm) required to achieve dark, uniaxial state in the above homeotropic cell.

to an arrangement wherein the polar directors in the neighbouring layers are at an angle of 90°. Similar transformations were noted in planar cell as well, wherein; this intermediate state possesses an intermediate birefringence value of 0.112, which lies exactly in between the birefringence values in the antiferroelectric and ferroelectric states.

3.2. *Dielectric Soft Mode*

Figure 3 shows the plot of induced polarization measured using the integral current technique for a 2 μm planar cell and 2nd harmonic response of transmittance with temperature measured for a homeotropic cell of 9 μm thickness. Both these physical parameters hint that during transition from $SmAP_R$ to $SmAP_A$ phase, the sample may undergo softening.

Further, to understand the behaviour of this phase transition, we studied the dielectric relaxation behaviour of this sample in a 10 μm planar cell as the sample is cooled from 165 °C to 60 °C. In the entire temperature range of measurements, the dielectric spectra shows two relaxation processes P2 (intermediate frequency) and P3 (high frequency) between 10 Hz to 100 MHz. When the dielectric loss spectra (ε'') are fitted to Cole-Cole equation the intermediate frequency peak P2 attracts attention. Figure 4(a) shows the temperature dependence of the dielectric amplitude ($\delta\varepsilon$) and the relaxation frequency f_R. Both $\delta\varepsilon$ and f_R diverge on approaching $SmAP_R$-$SmAP_A$ phase transition similar to observed for SmA- $SmAP_A$ phase transition. When this dielectric amplitude ($\delta\varepsilon$) and the electro-optic response (EC_2) with temperature are fitted to the power law $k/(T-T_C)^{\gamma}$ equation with γ as exponent; γ varied from 1 to 1.35 in case of the dielectric response depending strongly on the fitting temperature range while γ for the electro-optic response is rather stable

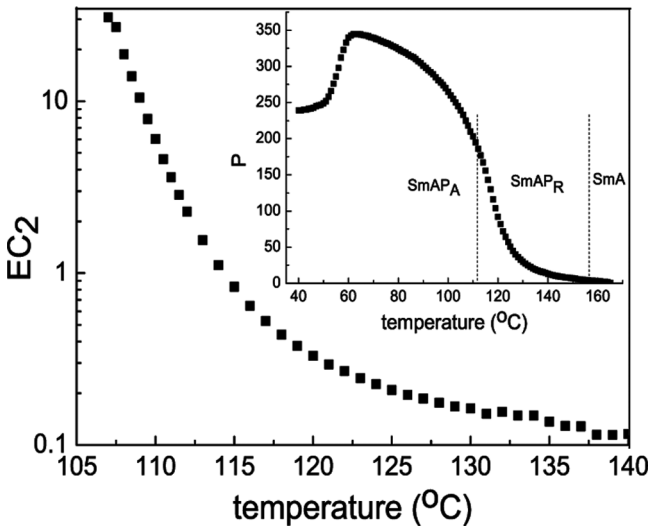

Figure 3. Temperature dependence of 2nd harmonic electro-optic response in a 9 μm homeotropic cell at 110 Hz. Inset: Temperature dependent electric field induced polarization in a 2 μm planar cell at $\mathbf{E} = 10\,V/\mu m$, 110 Hz.

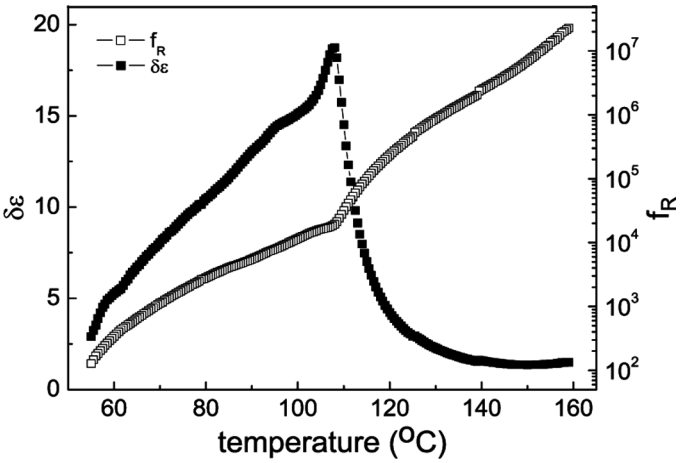

Figure 4. Dielectric responses in 10 μm planar cell. $\delta\varepsilon$ (dielectric amplitude) and f_R (relaxation frequency) of process P2 with temperature.

with an approximate value of ~1.3. These results lead us to conclude that response of the polar directors in $SmAP_R$ phase on approaching the $SmAP_A$ transition behaves similar to the softening process observed before [11].

4. Conclusion and Perspectives

In summary, we have studied the electric field induced textural transformations in orthogonal biaxial smectic phase and have been explained as transformation from antiferroelectric to ferroelectric transition via an intermediate state. These transformations could be exploited to demonstrate liquid crystal display modes of high efficiency. We have also presented results which show that the material undergoes softening during transition from an orthogonal smectic polar random phase to an orthogonal smectic polar biaxial phase.

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